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Assessment of Radionuclide Concentrations in Tea Samples Cultivated in Chittagong Region, Bangladesh

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Abstract:
The purpose of the present study is to assess the activity concentration of radionuclides in tea samples which were cultivated in Chitttagong region in Bangladesh. Total thirteen (13) tea samples were collected from different tea garden of Fatickchari, Rangunia and Banshkhali upozilla in Chittagong. The activity concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in thirteen tea samples were measured, using a gamma spectrometry system with multichannel analyzer. The observed activity concentrations of ^{238}U found in tea samples varied from 2.4 ± 0.4 $Bq.kg^{-1}$ to 18.93 ± 1.72 $Bq.kg^{-1}$ with mean concentration 5.34 ± 0.38 $Bq.kg^{-1}$. The observed activity concentration of ^{232}Th found in the tea sample varied 1.4 ± 0.56 $Bq.kg^{-1}$ to 27.22 ± 3.65 $Bq.kg^{-1}$ with mean concentration 10.07± 0.83 $Bq.kg^{-1}$. The ^{40}K activity in these tea samples varied from 151 ± 18.72 $Bq.kg^{-1}$ to 1243 ± 83.91 $Bq.kg^{-1}$ with mean concentration 430 ± 35.5 $Bq.kg^{-1}$. No ^{137}Cs activity was detected at any of the samples in the whole study area. The calculated values of Radium equivalent activity (Ra_{eq}) and the internal hazard index (H_{in}) for the tea samples are lower than the international permissible value of unity UNSCEAR (2000). It was concluded that the activity level of tea samples in the present study was non-threatening to public health.

Key words: Tea sample, Activity concentration, Gamma Spectrometry system, Radium equivalent activity (Ra_{eq}) and the internal hazard index (H_{in}).

Introduction

Tea is one of the most popular beverages all over the world which is prepared from the leaves of a shrub camellia sinensis. Green and black teas are the two most popular types. Drying and roasting the leaves produces green tea, black tea is obtained after a fermentation process. Economic and social interest in tea is clear from the fact that about 18–20 billion tea cups are consumed daily in the world [1]. Bangladesh is an important tea producing country. Its tea industry dates back to British rule, when the East India Company initiated the tea trade in Chittagong in 1840. The industry accounts for 3% of global tea production, and employs more than 4 million people. Tea has been one of the major exportable items of Bangladesh since 1971. Chittagong is the birthplace of the Bangladesh tea industry. The tea is grown in the northern and eastern districts, the highlands, temperate climate, humidity and heavy rainfall within these districts provide a favorable ground for the production of high-quality tea. European traders established the first sub continental tea garden in the port city of Chittagong in 1840, when plantations were set up beside the Chittagong Club using Chinese tea plants from the Calcutta Botanical Garden [2].

The medicinal value of tea for prevention and treatment of many health problems has

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become more and more commonly known [3] Tea contains flavonoids, minerals and trace elements that are essential to human health. Environmental radiation originates from a number of naturally occurring and man-made sources. The largest proportion of human exposure to radiation comes from natural sources of external radiation, including cosmic and terrestrial radiation and from inhalation or ingestion of natural radioactive materials. The United Nations Scientific Committee on the Effects of Atomic Radiation has estimated that exposure to natural sources contributes >70% of the population radiation dose and the global average human exposure from natural sources is 2.4 mSvy-1 (cosmic ray 0.4, terrestrial gamma ray 0.5, radon 1.2, and food and drinking water 0.3[4]. Acute health effects of radiation, appearing with symptoms of nausea vomiting, diarrhea, weakness, headache, anorexia leading to reduced blood cell counts and in very severe cases to death, occur at high doses of exposure of the whole body or large part of the body. Therefore, acute health effects of radiation are practically not a concern for continuously monitored for radioactivity content—central drinking water supplies. However, extreme situations of possible terrorist use of radioactive materials to contaminate drinking water supplies, theoretically, cannot be excluded [5]. Environmental radionuclides are concentrated in tea leaves and consumption of black tea as a beverage may contribute to an internal radiation dose[6]. Measurement of the concentrations of radionuclides present in black tea permits the assessment of the dose received by intake of this beverage. Radiation levels of the samples can be determined by a gamma spectrometer, equipped with a high-purity germanium detector. The main objective of this study was to quantify the presence of natural radionuclides i.e.^{238}U, ^{232}Th and ^{40}K in tea samples cultivated in Chittagong and to estimate the potential biological hazards by calculating the radium equivalent index, Ra_{eq} and hazard index, HI for consumers. The obtained data could be used as the baseline for future reference.

Materials and Method

Area of Study:

A total number of 13 tea samples were collected from various tea gardens of three different locations namely Fatickchari, Rangunia and Banshkhali upozilla in Chittagong. All the samples were collected during 2017. The sampling stations are located by the geographical position of Latitude ranged over 22°02′55″N - 22°45'23" N and Longitude 91° 42' 57" - 91°56′30″E. Today Bangladesh has 163 commercial tea estates including many of the world's largest working plantations [7] . Tea production in Bangladesh is about 53million kgyear^{-1}.There are 17 tea gardens are in Fatickchari Upazila. In Rangunia there are three tea gardens is situated. These are Agunia, Kodala and Thandachhari tea garden. Chandpur-Belgaon tea garden is situated in Banshkali. The Port of Chittagong is the largest international seaport on the Bay of Bengal and the Chittagong Tea Auction sets the price of Bangladesh Tea. This is why the tea estates of Chittagong were chosen for this study. The map of Chittagong district, Fatickchari, Rangunia and Banshkali Upazila are shown in fig-1.

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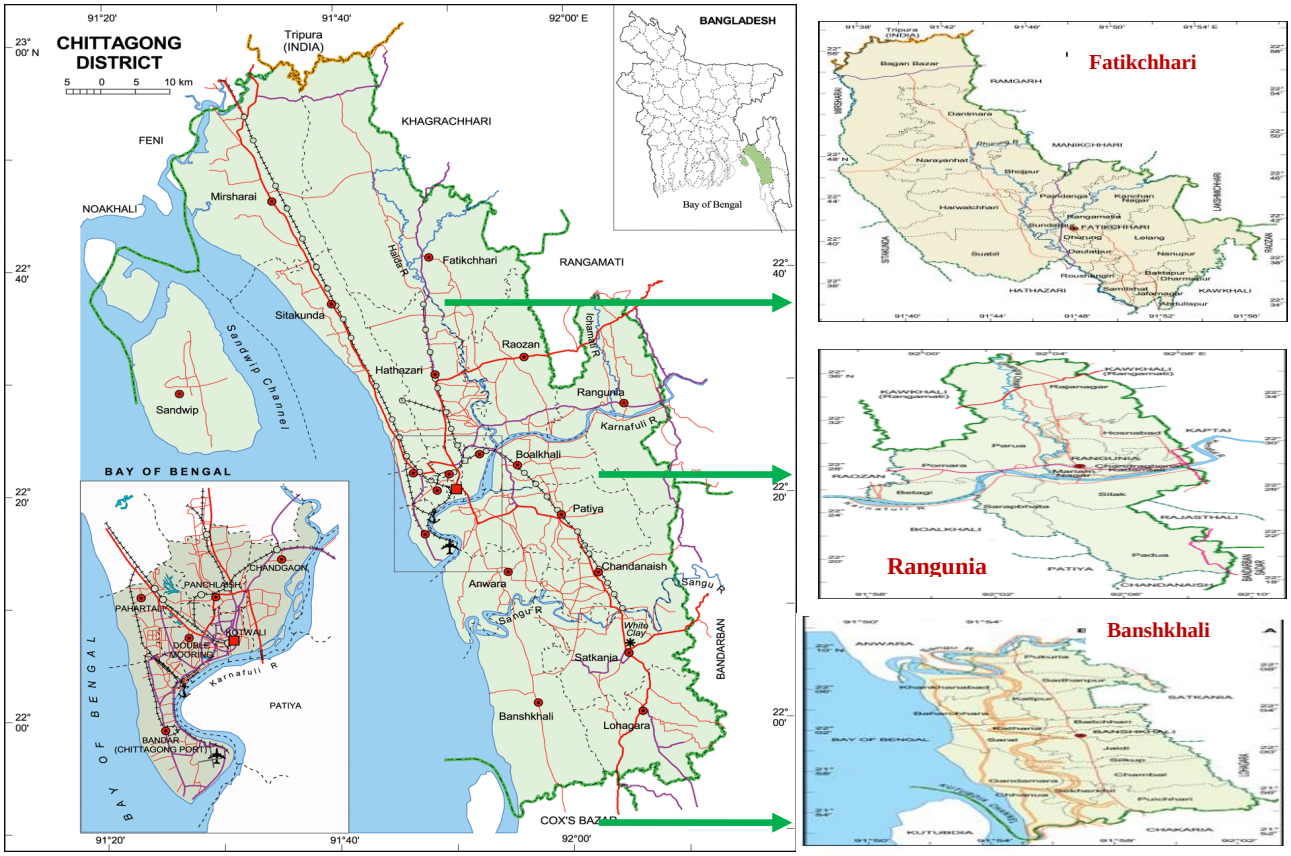

Fig-1: Map of Chittagong district, Fatickchari, Rangunia and Banshkali Upazila

Sample collection and preparation

In this study, total thirteen (13) tea samples were collected from different tea gardens from three different areas namely Fatickchari, Rangunia and Banshkhali upazilla in Chittagong to investigate the natural radioactive concentration using gamma spectrometry. After collecting the samples, each one was put in a plastic bag and given a label to show its name. After collecting the samples, all the tea samples were brought into the laboratory. In order to remove moisture, tea samples were dried in a drying oven at 80°C until constant mass was obtained. The samples were ground and homogenized in the laboratory. Adequate samples were put into cylindrical plastic analysis containers to reach a geometric homogeneity all around the detector. Plastic analysis containers had a 6.5 cm diameter and 7.5 cm height. Then samples were weighed using a high sensitive digital weighting balance and sealed with paraflim to prevent the escape of radon gas. Samples were kept for 30 days for secular equilibrium of ^{226}Ra decay products before the measurements. Then each sample was measured and the values were given in Bq. kg^{-1} dry weight.

Radioactivity Measurement

To qualitatively identify the contents of radionuclides in both regions soil sample and to quantitatively determine their activities, all prepared samples were measured by means of gamma-ray spectrometry system using ORTEC high purity Ge-detector for 15000 sec.

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The equal counting time for background and sample measurement was chosen to minimize the uncertainty in the net counts. The spectrum of each sample was analyzed and the identification of unknown radionuclides was carried out by considering their peak centroid energies. The centroid energies of the peaks from the spectrum were compared with the reference gamma-ray energies obtained from the literature. Before the measurements, energy calibration was done by using peaks of ^{137}Cs and ^{60}Co radionuclides of the standard point radioactive source. The number of counts under the full-energy peak areas (corrected for background peak areas), the counting time, the absolute full-energy peak efficiency for the energy of interest and the gamma-ray emission probability corresponding to the peak energy are used for the calculation of the activity concentration of a particular radionuclide in the measured samples. One problem with the direct determination of the activity of ^{238}U and ^{232}Th is due to the low relative gamma-ray intensities following their decay. However, in a state of secular equilibrium, the activity of ^{238}U and ^{232}Th can be estimated through several intensive gamma-ray lines of their daughter products in the decay chains. The activity concentrations of ^{238}U and ^{232}Th were determined from the average concentration of nuclides [Pb^{214} (295.2keV), Pb^{214} (351.9keV), Bi^{214} (609.3keV) and Bi^{214} (1120.2keV)] and [Pb^{212} (238.6keV), Tl^{208} (583.1keV), and Ac^{228} (911.2keV), Ac^{228} (968.9keV)] respectively. The activity concentrations of ^{40}K were determined directly by measurement of the gamma-ray transitions at 1460.8keV. The efficiency calibration of gamma spectrometry system was performed using Eu-152 standard volume source. The efficiency calibration source had vegetation matrix and 10 radionuclides that have the energy range of 121–1407 keV. Fig.-2 shows the spectrum of gamma-rays from a tea sample of Chittagong.

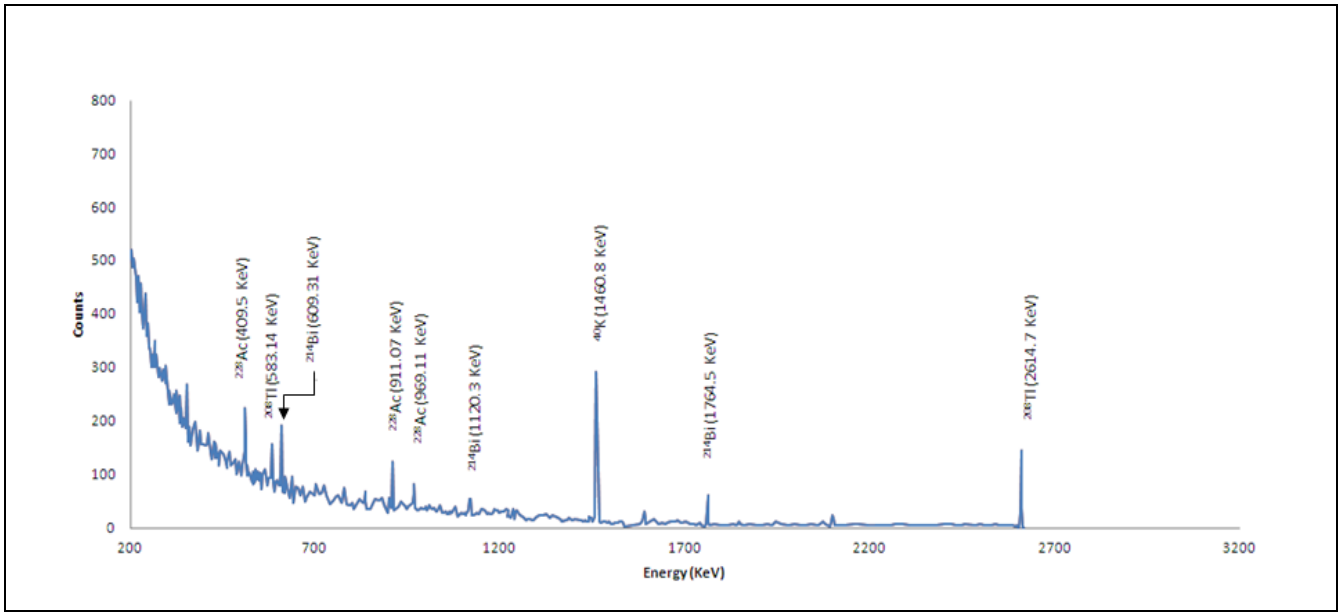

Fig.2: Spectrum of the gamma-rays from a tea sample of Chittagong.

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Data Analysis

Before the measurements an empty plastic sample container was counted in the same manner as the samples for the determination of the background effects. The specific activity, in terms of the activity concentration, is defined as the activity per unit mass of the sample. The specific activity of individual radionuclides in water samples is given by the following equation:

$$A = \frac{N \times 100 \times 1000}{P\gamma \times \varepsilon \times W}$$

----------(1)

Where, N is Net counts per second; Pγ is

Transition probability of gamma ray or Branching ratio; ε is efficiency in percent and

W is Weight of the sample in kg.

The term net count rate associated with the activity measurement is the difference between the gross count rate of the sample (which is the summation of background count rate and sample count rate) and the background count rate. Each count rate includes standard deviation and the standard deviation of the net count rate can be expressed as,

$$\sigma = \pm \sqrt{\frac{A_s}{T_s} + \frac{A_b}{T_B}}$$ ----------

----------(2)

Where, A_s is sample count rate in cps, A_b is background count rate in cps, T_s and T_b are counting time of sample and background. The standard deviation is also a measure of the dispersion of a collection of numbers. It can apply to a probability distribution, a random variable, a population or a data set. The standard deviation is usually denoted with the letter **σ.** The value of σ then converted into error activity in the unit of Bq.kg^{-1} as well as main activity by using activity equation.

Dose Estimation

Radium equivalent activity (Ra_{eq}) is utilized to evaluate the risks of materials that contain ^{238}U, ^{232}Th and ^{40}K in Bq.kg^{-1} [8], which is identified by presuming that 185 Bq.kg^{-1} of ^{238}U or 260 Bq.kg^{-1} of ^{232}Th or 4810 Bq.kg^{-1} of ^{40}K produce the same gamma dose rate. The Ra_{eq} of a sample in (Bq.kg^{-1}) can be achieved using the following relation [9, 10]:

$$Ra_{eq} = A_U + (1.43 A_{Th}) + (0.077 A_K)$$ ---------

----------(3)

The internal hazard index can be quantified by the internal hazard index (H_{in}). This is given by the following equation[9,10]:

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}$$ ----------

----------(5)

The internal hazard index has to be less than one as well to provide safe radionuclide levels in tea.

Results and Discussion:

Measurement of Activity concentration

Measurement of radionuclide concentration in foods and beverages is of great significance for the protection of human health. The activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in the tea samples, collected from different parts of the studied areas, are presented in Table 1. The concentrations of the radioisotopes in tea samples are reported in Bq.kg^{-1} of dry weight. To determine the concentrations of radionuclides in the samples, Equation 1 was used.

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Table-1: Activity concentrations (Bq.kg^{-1}) of radionuclides in tea samples (mean±SD), Radium equivalent activity (Ra_{eq}) and internal hazard index (H_{in}).

Sample code	Name of tea sample	^{238}U	^{232}Th	^{40}K	Ra_{eq}	H_{in}
Tea-1	Aasia	18.93± 1.72	17.6 ± 1.06	237± 74.6	62.36	0.219
Tea-2	Neptune & Andarmanik	5.61± 1.81	7.18 ± 1.20	378± 79.88	45.00	0.136
Tea-3	Dantmara & Naseha	3.42 ± 0.21	11.96 ± 1.93	151 ± 18.72	32.14	0.096
Tea-4	Haldavalley	8.29± 1.38	5.27 ± 0.85	167± 68.39	28.68	0.099
Tea-5	Kaiyacherra Dalu	2.47± 0.68	23 ± 5.44	200± 58.46	50.75	0.143
Tea-6	Kornafuli	3.57± 1.42	4.18 ± 0.62	277 ± 60.02	30.89	0.093
Tea-7	Panchabati	3.76± 1.08	1.4 ± 0.56	384 ± 70.72	35.34	0.105
Tea-8	Rangapani & Baramasia	4.41± 2.97	3.79 ± 0.26	609 ± 69.62	56.71	0.165
Tea-9	Oodaleah	4.12 ± 1.86	4.39± 1.19	226 ± 66.03	27.80	0.086
Tea-10	Ramgarh	3.2 ± 2.18	4.65 ± 1.76	625 ± 62.37	57.98	0.165
Tea-11	Chandpur-Belgaon	5.67 ± 2.16	12.41 ± 2.82	380 ± 62.06	52.64	0.157
Tea-12	Thandacheri	2.4 ± 0.4	9.16 ± 1.26	712 ± 68.84	70.29	0.196
Tea-13	Kodala	3.56 ± 0.69	27.22 ± 3.65	1243± 83.91	138	0.382

The ranges of activity concentration and average activity concentration of different radionuclides in the collected tea samples are shown in Table-6.4. The observed activity concentrations of ^{238}U found in tea samples varied from 2.4 Bq.kg^{-1}to 18.93 Bq.kg-1with average activity concentrations 5.34 ± 0.38 Bq.kg^{-1}. Sample Tea-1 has highest value of the ^{238}U activity concentration. Sample Tea-12 has smallest value of the ^{238}U activity concentration. The observed activity concentration of ^{232}Th found in the tea sample varied 1.4 Bq.kg^{-1} to 27.22 Bq.kg^{-1} with mean concentration 10.07± 0.83 Bq.kg^{-1}. Sample Tea-13 has highest value of the ^{232}Th activity concentration. Sample Tea-7 has smallest value of the ^{232}Th activity concentration. The activity concentrations of ^{238}U and ^{232}Th for the tea samples are shown graphically in Fig.3.

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Table 2: Ranges of activity concentration and average activity concentration of ^{238}U, ^{232}Th and ^{40}K in the collected thirteen tea samples

Radionuclides	Range of activity concentration ($Bq.kg^{-1}$)	Average activity concentration ($Bq.kg^{-1}$)
^{238}U	2.4 - 18.93	5.34 ± 0.38
^{232}Th	1.4 - 27.22	10.07 ± 0.83
^{40}K	151 - 1243	430 ± 35.5

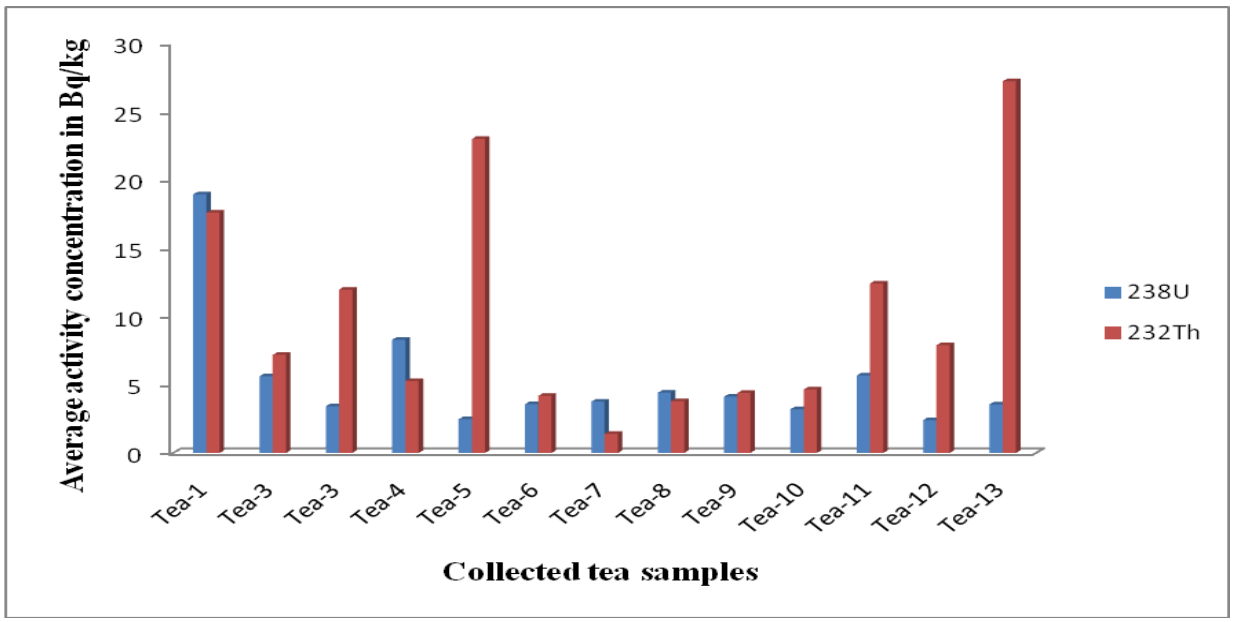

Fig. 3: The activity concentrations of ^{238}U and ^{232}Th for the tea samples.

The activity concentrations of ^{40}K in these tea samples range from 151 to 1243 $Bq.kg^{-1}$ with mean concentration 430 ± 35.5 $Bq.kg^{-1}$. Sample Tea-3 has smallest value of the ^{40}K activity concentration. Sample Tea-13 has highest value of the ^{40}K activity concentration. The activity concentrations of ^{40}K for the tea samples are shown graphically in Fig.4.

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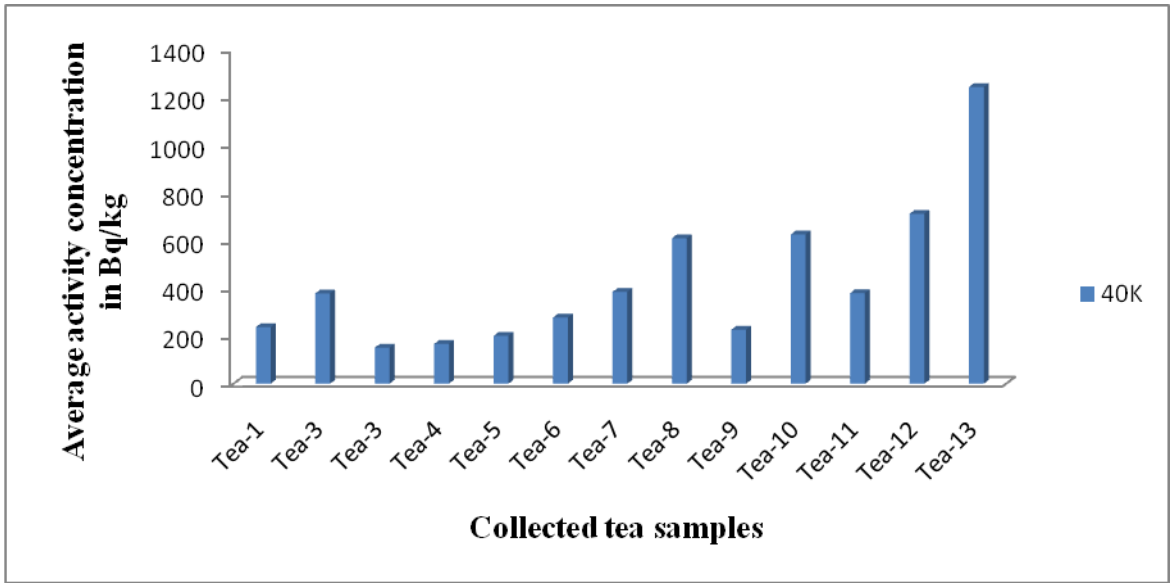

Fig. 4: The activity concentrations of ^{40}K for the tea samples

Activity concentration of artificial radionuclide such as ^{137}Cs was detected at any of the samples in the whole study. Based on the findings, the activity concentration of ^{40}K has significantly higher than other radionuclides in all the collected samples as shown graphically in Table-1. This may be due to the high concentration of ^{40}K and higher transfer factor of the radionuclide because of the specific metabolic processes involving potassium in plants. Also, the extra use of Murate of Potassium fertilizer may be another factor causing the increase of ^{40}K concentration in the tea samples. The present findings were compared with the results reported in previous studies. The average activity concentrations of ^{238}U, ^{232}Th and ^{40}K for tea samples in the present study along with the values of other countries are presented in Table 3. The activity concentrations of ^{40}K in various tea samples of different countries are shown in fig.-5.

Table 3: Average activity concentrations of ^{238}U, ^{232}Th and ^{40}K in tea samples for different countries with that of the present work

Countries	Average activity concentrations in Bq.kg^{-1}			References
	^{238}U	^{232}Th	^{40}K	
Egypt	16	3	623	[11]
Iran	_	0.037	410	[12]
Turkey-1(Karadeniz region)	_	_	485.4	[13]
Turkey-2(Market tea)	0.9	2.7	501	[14]
Serbia-1(Market tea)	0.6-8.2	1.7-15.1	126-1243.7	[15]
Serbia-2(Mountain region)	_	_	130-1160	[16]
Bangladesh (Present study)	5.34	10.07	429.91	

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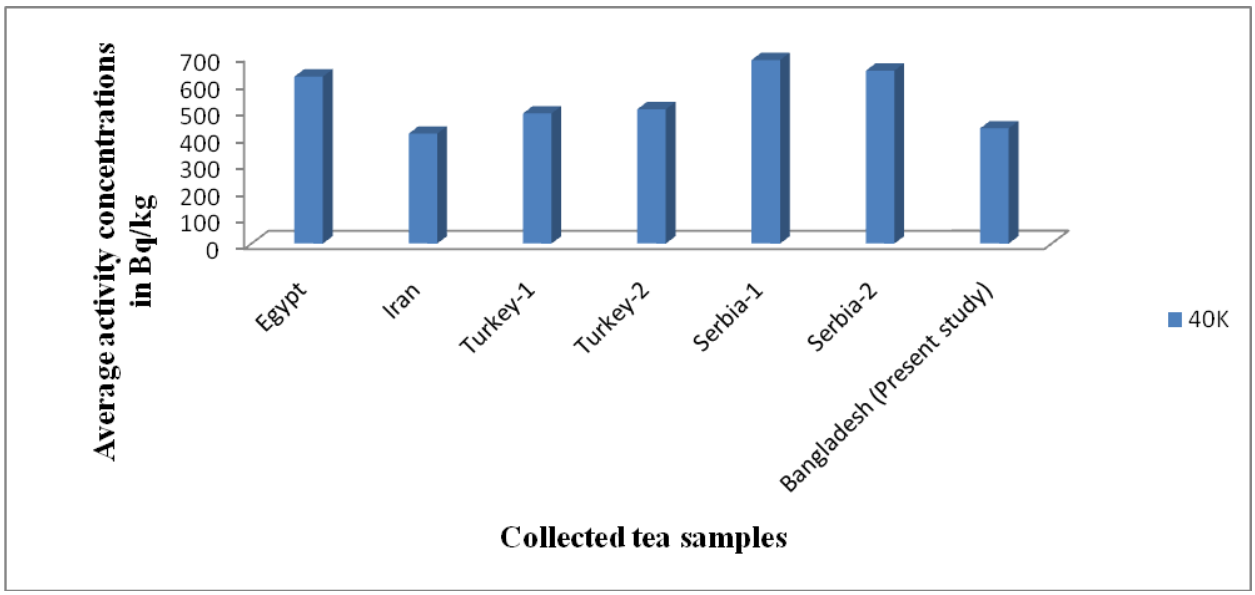

Fig 5: Activity concentrations of ^{40}K in various tea samples of different countries

In the present study the average activity concentration of ^{238}U is 5.34 Bq.kg^{-1}which is lower than the value of Egypt. This value is higher than the value of Turkey-2 and within the range of the value of Serbia-1.The average activity concentration of ^{238}Th in present study is 10.07 Bq.kg^{-1} which is higher than the values of Egypt, Iran and Turkey-2. This value is within the range of the value of Serbia-1.In the present study the average activity concentration of ^{40}K is 429.91 Bq.kg^{-1} which is lower than the value of Egypt, Turkey-1 and Turkey-2. This value is higher than the value of Iran and within the range of the values of Serbia-1 and Serbia-2. Humans are exposed to both external and internal radiations from radionuclide sources [17]. ^{40}K, ^{238}U, ^{232}Th and their daughters are the main contributors to the internal radiation dose received by man from the natural sources. This internal radiation dose varies with the radionuclide concentration in foodstuff and the rate of intake, which differs from one group of people to another. Table 1 presents the calculated values of Radium equivalent activity (Ra_{eq}) and the internal hazard index (H_{in}) for the tea samples. The results showed that Ra_{eq} ranged between 27.80 and 138 Bq.kg^{-1}, and the average Raeq value was reported to be 52.97 Bq.kg^{-1}. The values for the radium equivalent activity (Ra_{eq}) are found to be within the world average allowed maximum value of 370 Bq.kg^{-1} UNSCEAR (2000) [4]. The H_{in} values varied from 0.086 to 0.382 in the samples. The average H_{in} value was reported to be 0.157. All values of internal hazard indices are lower than the international permissible value of unity UNSCEAR (2000) [4]. As presented in Table 1, the current findings indicated that the tea samples were free from radiation hazards. It was concluded that the activity level of tea samples in the present study was non-threatening to public health.

Conclusion

In this study, the concentrations of ^{238}U, ^{232}Th and ^{40}K in tea samples cultivated in Chittagong, Bangladesh were determined, using gamma spectrometry. The tea samples from Chittagong did not contain any

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anthropogenic radionuclide (^{137}Cs), but the natural radionuclides such as ^{238}U, ^{232}Th and ^{40}K were identified. The values for the radium equivalent activity (Ra_{eq}) are turned to be within the international average allowed maximum value of 370 Bq.kg-1. The activity concentrations of radionuclides were lower than the standard limits, established by international organizations. This study could be of help as a data base for radionuclide concentration and radium equivalent activity. The value of hazard internal is lower than the international permissible value of unity. In general terms, it can be concluded that the implemented technique show good results when matched with other literature data. Also it can be concluded that samples under study, which have been analyzed, are safe for human consumption because their radioactivity levels are less than the maximum permitted level.

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